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Preparation of wood plastic composite sheets by lateral extrusion of solid woods using their fluidity

Tsunehisa Miki*, Masako Seki, Soichi Tanaka,
Nobuo Sobue, Ichinori Shigematsu, Kozo Kanayama

*National Institute of Advanced Industrial Science and Technology (AIST),
2266-98 Anagahora, Shimo-shidami, Moriyama-ku, Nagoya 463-8560, Japan*

Abstract

During the course of our studies, we have learned that solid wood has significant deformability characteristics under specific conditions, resulting from slippage occurring among wood cells when wood treated with small-molecular resins is compressed under heat. Furthermore, the deformed shapes obtained are maintained when the resin introduced into the wood cells is cured. In this study, the lateral extrusion of thin discs of wood treated with thermosetting phenol resin is performed to apply this flowlike deformation behavior of wood to wood-forming techniques. Resin-treated thin discs of wood placed in a heating container were compressed by a punch, and under constant pressure the wood flowed into the cavity of a mold. Successful molding conditions were achieved by varying the pressure, temperature and wood fiber direction for the extrusion ratio of 42. The mechanical properties of the wood plastic composite (WPC) sheets obtained by this extrusion process are affected by the fiber direction of the wood in the extrudate, which is almost equivalent to the longitudinal direction of the cellulose crystals of wood. The maximum bending strength of the WPC sheets obtained was about 180 MPa, which is suitable for engineering plastics.

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* Corresponding author. Tel.: +81-75-736-7527; fax: +81-75-736-7533.

E-mail address: tsune-miki@aist.go.jp

1. Introduction

Wood is a sustainable and renewable resource that can be used as a material as well as a fuel. Compared to underground resources, wood resources exist all over the face of the planet and are easily prepared for use by processes such as cutting and drying. Furthermore, wood is produced by photosynthesis of the tree, by which atmospheric carbon dioxide (CO₂) is adsorbed and transformed into carbohydrates required for making wood components and structures. This indicates that the promotion of the use of wood and the long-term use of wood and wood-based materials will lead to a reduction of CO₂ emissions as they fix CO₂ in their component parts. Therefore, development of advanced applications for wood-based materials is of interest in the fields of engineering, agricultural and materials science.

Against this social background, wood-plastic composites (WPCs) and natural fiber-reinforced plastics (NFRPs), produced by compounding small wood/plant elements with thermoplastic polymers, have seen a rapidly growing market this decade, as shown by Asta Eder & Michael Carus. These wood-plastic composites offer several advantages, such as enhancement of specific properties such as stiffness and thermal behavior, reduced price of the material and improved recyclability when compared to traditional glass fiber-reinforced plastics. Their applications have been expanding from outdoor nonstructural members to indoor components, which represents severe environment conditions associated with water for normal wood. As the waterproofness of WPCs depends heavily upon the polymers used, the polymer content in composites has reached almost 50 %. Therefore, the affinity of wood for a polymer and the covering of a wood element with polymer is of significance in maintaining the properties of a composite. Many attempts have been made to improve the affinity between hydrophilic wood elements and hydrophobic polymers by compatibilizing polymers having hydroxyl groups or hydrophobized wood. Hydros swelling of a WPC using a compatibilizer, however, gradually occurs in humid conditions, although the strength properties improve greatly. This is because the improved interfacial property works on the surface of wood elements in a composite, and the initial properties of wood elements hardly change. To improve the properties of wood itself, a polymer should interact with wood components such as noncrystalline substances like hemicellulose and lignin. It is thus necessary for polymers to enter the walls of wood cells where the intrusion of molecules is limited by weight to a molecular size of less than 500 (Ishimaru et al., 1999). Because polypropylenes and polyethylenes, which have molecular sizes greater than 10,000, are mainly used in compounding-type WPCs, the properties of wood are assumed not to change. Other ways to improve the properties of WPCs in the presence of water are the hydrophobization of wood elements such as by using acetalization, thermal or thermalhydro treatments. These treatments are effective ways for improving the water resistance, but they deteriorate the mechanical properties of wood. Problems have been noted in compounding WPCs and NFRPs in terms of their material properties in the presence of water in addition to problems with practical production processes.

To resolve these issues, we have developed completely different WPCs from the compounding types in which both the deformability and physical properties of solid wood are modified by the interaction of polymers with wood components (Miki et al., 2014). Polymers existing in wood act as plasticizers as well as binders under certain temperature conditions, and wood containing polymers shows a flowlike deformation behavior under compression. In this technique, conventional plastic-forming processes for metallic materials using a mold and pressing machine are applied for shaping wood, by which the speed and productivity of forming is drastically improved in comparison to conventional wood-shaping techniques such as carving and extruded WPCs. Furthermore, our newly developed WPC exhibit either a thermoplastic or thermosetting nature depending on the polymers introduced into the wood cells.

In this basic study, WPC sheets are prepared by the lateral extrusion of solid wood modified with a thermosetting phenolic resin based on our techniques. To investigate the effect of extrusion conditions such as temperature and pressure, the effects of resin introduction on thermal behavior related to softening and hardening of modified solid wood are measured. The possibilities and problems in the application of WPCs also are discussed.

experiment. The mold consists of three main parts; a punch, an upper plate, and a base plate. The base plate has a 2-mm deep concave section that functions as a cavity when the upper plate is installed, as seen in the initial setup condition. The mold is heated by cartridge heaters and the heating plates of a pressing machine so as to achieve the target temperature with a maximum variation of $\pm 3^\circ\text{C}$ on the cavity surface. The contact surface of the cavity has a mirror (specular) finish and before each extrusion run, a mold release agent containing silicone was sprayed onto the surface of the mold.

As wood is a natural orthotropic material that grows from the stem direction—namely longitudinal, radius and tangential—the orientation of the wood samples was also considered. In this experiment, two sample arrangements were used: (i) all tangential directions, and (ii) all longitudinal directions, along the extrusion direction.

Figure 1 (right-hand side) shows an example of changes in the experimental parameters for the punch load and stroke. In the initial setup condition, direction-arranged samples weighing 22–25 grams were placed in the container of the mold, which had been preheated to the target temperature T . At the beginning of the process, the samples were compressed to the target pressure P . After reaching this pressure, the stroke was regulated to maintain the target load during the pressure-holding process. During the pressure-holding time of three minutes, the stroke was gradually increased as the samples flowed and extruded into the cavity. The zero point of the stroke was the displacement where the sample thickness became 2 mm, and this zero-stroke—defined as the difference from the final stroke s , illustrated in the figure—is discussed in the results. Finally, the extruded product was removed after the mold had cooled.

3. Result and discussion

3.1. Deformation and flow behavior in extrusion process

From the results of thermal analysis, the extrusion temperature of the WPC product was selected between 120 to 170 $^\circ\text{C}$ as it is necessary for a thermosetting wood to flow in a softened state, as well as to improve the strength of products after extrusion (Miki et al., 2012). Fig. 2(a), (b) and (c) show the stroke changes indicated by the difference from the final stroke s during the pressure-holding process at an extrusion pressure of 100 MPa. As seen in Fig. 2(a), the difference from the final stroke did not reach zero when untreated wood with an MC of 10% was compressed—even if the extrusion temperature was set higher. This means that untreated wood did not flow and extrude into the cavity due to insufficient fluidity. When wood impregnated with PF resin with an MC of 10% was compressed, the stroke changed drastically. It can be seen in Fig. 2(b) that the stroke reached zero at all temperatures. As the PF-treated wood had improved fluidity, the extrusion run was performed to completion and this lead to increased wood filling in the cavity. Higher extrusion temperatures resulted in a faster run and filling rate, and, especially at temperatures above 150 $^\circ\text{C}$, full extrusion was achieved within a minute. The fiber direction in the extrusion, determined by the arrangement of wood in the initial setup condition, affected the extrusion behavior. Because a longitudinal wood flow required a higher pressure due to the lower fluidity compared to a tangential flow, the higher temperature resulted in insufficient extrusion (Yamashita et al., 2009).

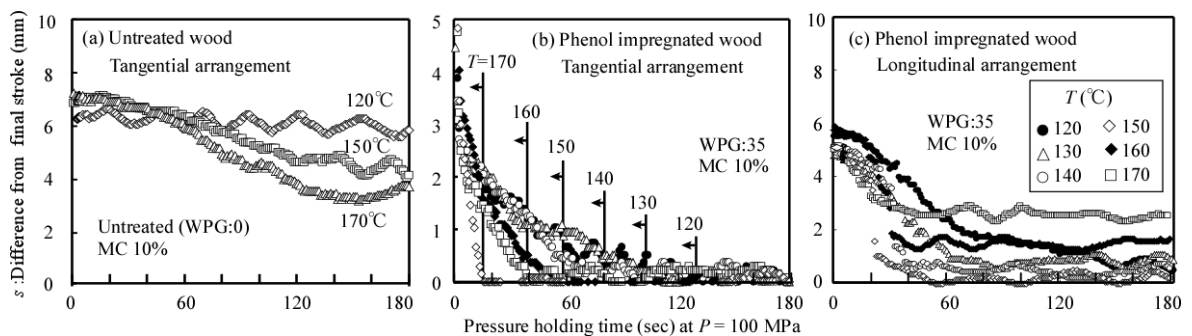


Fig. 2. Flow behaviour of solid wood during the extrusion process at different temperatures.

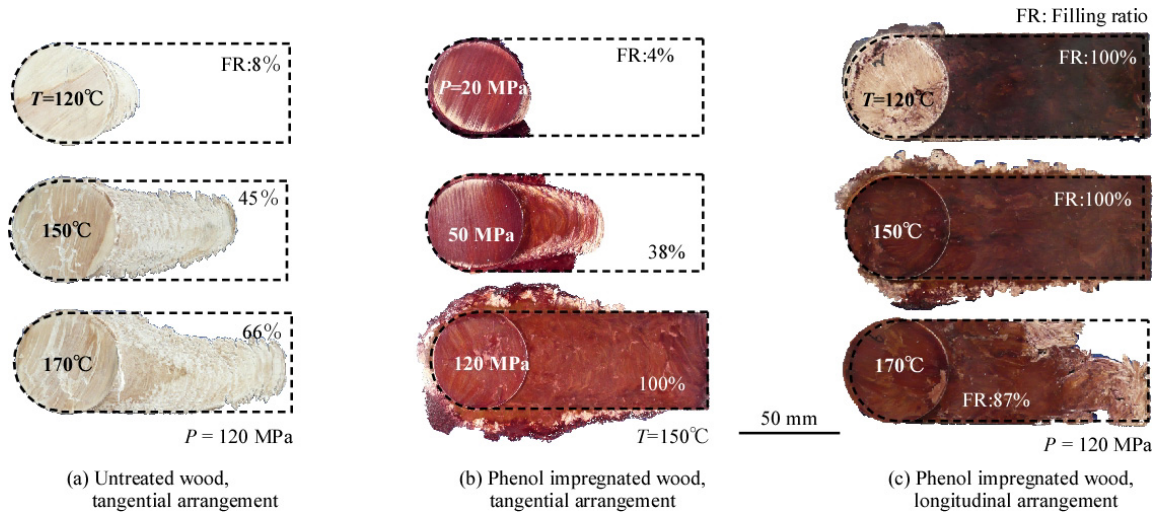


Fig. 3. Cavity-filling states of solid wood during the extrusion process.

3.2. Cavity filling states in extrusion process

Fig. 3(a), (b) and (c) shows the filling states of wood extruded at various mold temperatures and pressures, which correspond to the conditions shown Fig. 2. For untreated wood, as shown in Fig. 3(a), complete filling could not be obtained. The filling ratio as calculated from the area of the cavity occupied by wood was at most 66%. At similar temperature and pressure conditions— $T = 150\text{ }^{\circ}\text{C}$, $P = 120\text{ MPa}$ —PF-treated wood showed a greater formability than untreated wood, as shown in Fig. 3(b). The figure shows that burr was generated due to the excessive pressure applied. The PF-treated wood started flowing at 20 MPa, and extruded after gradual filling in the widthwise direction of the cavity with an increase in pressure. It can be seen in Fig. 3(c) that filling of the cavity was not completed when extrusion with a longitudinal arrangement of wood was conducted at a higher temperature, with the filling ratio dropping to 87% when the extrusion temperature was increased to $170\text{ }^{\circ}\text{C}$ from $150\text{ }^{\circ}\text{C}$.

3.3. Mechanical properties of wood plastic composites

Fig. 4 shows the effects of the extrusion temperature and wood arrangement on the bending strength and bulk density noted in the parentheses. The wood arrangement and bending specimens cut from an extrudate are shown in the figure. It was found that the extrudate produced in the tangential arrangement showed a higher strength than that produced in the longitudinal arrangement. The dispersion of bending strength in the tangential extrusion, however, is greater than that in the longitudinal direction. While in the tangential arrangement extrusion the strength tends to decrease along toward to the tip, such a tendency was not seen in the longitudinal arrangement. The bulk density of the extrudate reached similar values, around 1.36 g/cm^3 , for both the tangential and longitudinal arrangements at all extrusion temperatures. However, the bending strengths for the tangential arrangement are twice those for the longitudinal arrangement. This enhancement of the bending strength appeared to be derived from the fiber orientation in the extrudate, which was confirmed by the X-ray diffractometry pole figures shown in Fig. 4. It was found that the extrudate obtained maximum strength at an extrusion temperature of $150\text{ }^{\circ}\text{C}$, where the peak temperature of phenol resin curing was detected in thermal analysis in previous studies.

As this thermosetting WPC extrusion can be classified as nonequilibrium processing, in which the temperature and moisture conditions change drastically during forming, it is considered that the true sample temperature will be different from the mold temperature. However, extrusion temperatures above $150\text{ }^{\circ}\text{C}$ seem to be too high to allow flow while maintaining PF curing at a lower level.

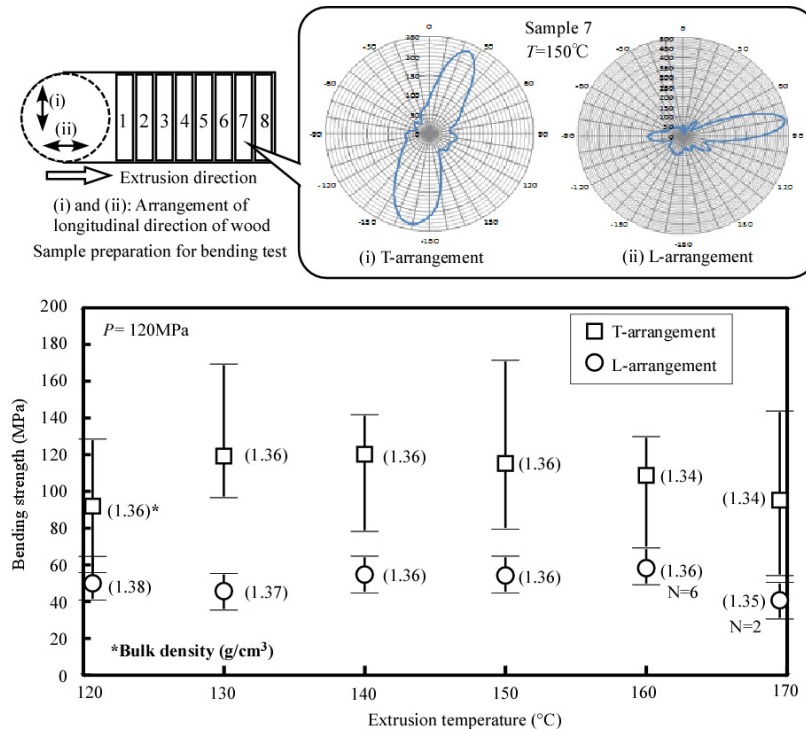


Fig. 4. Mechanical properties of WPC product and fiber orientations in products.

4. Conclusion

As a novel wood-shaping technique—as opposed to removal and deformation processes—we proposed a new processing method for thermosetting wood plastic composites, which originated from cell-cell slippage. By introducing low molecular weight phenol resin into wood, solid wood could be induced to flow easily so that a lateral-type extrusion can be applied for wood. By controlling the temperature and fiber arrangements, strength properties similar to those of industrial engineering plastics such as acrylonitrile butadiene styrene, polyvinyl chloride, and polycarbonate could be attained for wood-plastic composites obtained by wood-flow forming. Compared to conventional wood-shaping techniques such as cutting, bending, compression, arbitrarily shaped products with improved mechanical properties could be obtained with higher productivity.

Acknowledgements

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References

- Asta Eder & Michael Carus, 2013, Global Trends in Wood-Plastic Composites. *Bioplastics MAGAZINE* 8, 16-17.
- Yamaguchi T., Ishimaru Y., Urakami H., 1999. Effect of Temperature on Dimensional Stability of Wood with Polyethylene Glycol II. Temperature dependence of PEG adsorption and mechanical properties of treated wood. *Mokuzaigakkaishi* 45, 441-447.
- Miki T., Sugimoto H., Shigematsu I., Kanayama K., 2014, Superplastic Deformation of Solid Wood by Slipping Cells at Sub-micrometre Intercellular Layers, *International Journal of Nanotechnology* 11, 509-519.
- Miki T., Seki M., Sugimoto H., Shigematsu I., Kanayama K., 2012, Mechanical Properties of Wood Plastic Composites Prepared by Wood Flow Forming Using Low Molecular Phenol Resin, *Proceedings of Biocomp 2012*, 154-161.
- Yamashita O., Yokochi H., Miki T., Kanayama K., 2009. The pliability of wood and its application to molding. *JOURNAL OF MATERIALS PROCESSING TECHNOLOGY* 209 (12-13): 5239-5244.